Synopsis

Magnetostrictive materials belong to an important class of smart magnetic materials which have potential applications as ultrasonic transducers, sensors, actuators, delay lines, energy harvesting devices etc. Although, magnetostrictive property is exhibited by almost all ferro and ferrimagnetic materials, the R-Fe type (R represents rare earth elements) intermetallic compounds display maximum promise owing to the large magnetostriction exhibited by them at ambient temperature. Among the several R-Fe type compounds, Tb-Fe and Sm-Fe alloys are found to exhibit maximum room temperature positive and negative magnetostriction respectively. Recently, Fe-Ga based alloys have gained significant interest as newly emerging magnetostrictive material due to a good combination of magnetic and mechanical properties. These magnetostrictive materials in thin film form are of interests for several researchers both from fundamental and applied perspectives. Currently, many researchers are exploring the possibility of using magnetostrictive thin films in micro- and nano-electromechanical systems (MEMS and NEMS).

Three material systems *viz.* Fe-Ga, Tb-Fe and Sm-Fe in thin film form have been chosen for our investigations. DC magnetron sputtering and e-beam evaporation techniques were used for deposition of these thin films on Si (100) substrates. Several aspects such as evolution of microstructure, film surface morphology, structure and change in film composition with different processing conditions were investigated in detail, as the existing literature could not provide a clear insight. Further, detailed magnetic characterizations of these films were carried out and established a process-structure-property correlation.

The thesis is divided into seven chapters. The first chapter presents a brief introduction of magnetostrictive phenomena and the physics behind its origin. A brief history of evolution of magnetostrictive materials with superior properties is also brought out. Introduction to the material systems considered for the present study has also been presented. Discussions on various aspects like crystal structures, magnetic properties, and phase diagrams of these material systems are also included in this chapter. Magnetostriction in thin films and its importance in current technological applications are discussed in short. Further, a summary of existing literature on thin films of these materials has been narrated to highlight the perspective of the work done in subsequent chapters. In addition to this, a clear picture of the grey area for further investigations has been provided. Formulation of detailed scope of work for this study is also provided in this chapter. Details of different experimental techniques used in this study for deposition and characterization of these films are given in chapter 2.

In the third chapter of the thesis a detailed study on the structural, microstructural and magnetic properties of Fe-Ga films deposited using dc magnetron sputtering technique are presented. The effect of sputtering parameters such as (i) Ar pressure, (ii) sputtering power, (iii) substrate temperature and (iv) deposition time/film thickness on the magnetic properties of the films are discussed in detail. All the films are found to be polycrystalline in nature with A2 type structure as evidenced from grazing incidence X-ray diffraction (GIXRD) and transmission electron microscope (TEM) studies. Surface morphology of the films are found to be affected with processing conditions considerably. Thermomagnetic behaviour of the films studied using a Superconducting QUantum Interference Device (SQUID) magnetometer under zero field cooled (ZFC) and field cooled (FC) conditions are also presented. The sputtering parameters are also found to influence the magnetic properties of the films through modifications in microstructure, surface morphology and film compositions. Irrespective of the sputtering parameters, room temperature (RT) deposited Fe-Ga films are found to exhibit large magnetic coercivity and large saturation magnetic field as compared to the bulk alloy of similar compositions which are not desirable for micromagnetic device applications. A significant improvement in the magnetic properties of the films was obtained in the films deposited at higher substrate temperatures and is correlated with modifications in grain size and film surface roughness. These films are also found to exhibit better magnetostriction than the RT deposited films. Further, the magnetic properties of Fe-Ga films as a function of film thickness in the range 2 – 480 nm are also presented. The nature of variation of coercivity with film thickness was correlated with grain size effect and explained successfully with the help of random anisotropy model.

In the fourth chapter, studies on the microstructural and magnetic properties of Tb-Fe films were presented. It was reported earlier that Tb_xFe_{100-x} films exhibit in-plane magnetic anisotropy for the films with x > 42 at.% of Tb and out-of-plane anisotropy for the composition 28 < x < 42. Presence of these anisotropies is technologically important for different applications. We have studied the magnetic properties of Tb-Fe films in these two composition range. Tb_xFe_{100-x} films with $54 \leq x \leq 59$ were prepared using dc magnetron sputtering technique under varying Ar pressure and sputtering power and the details about microstructural and magnetic properties are presented in this chapter. All the films are found to be amorphous in nature. While the composition of the film is found to remain constant with sputtering power, the Fe concentration in the film is found to be depleted with increase in Ar pressure. Magnetic properties are found to change from superparamagnetic to ferromagnetic behaviour with increase in sputtering power. Curie temperature of the films are found to be low (below RT) and is explained based on sperimagnetic ordering of magnetic sub-lattices.

The perpendicular magnetic anisotropy (PMA) or out-of-plane anisotropy behaviour of Tb-Fe films were not studied in detail as a function of film thickness. We have successfully prepared Tb_xFe_{100-x} films with $29 \leq x \leq 40$ using e-beam evaporation technique using alloy target composition of TbFe in order to study the PMA behaviour as a function of film thickness. The thickness of the films was varied from 50 to 800 nm. All the films are found to be amorphous and columnar growth structure with fine channels of voids are observed from the TEM studies. Detailed magnetization and thermomagnetic measurements were carried out using SQUID magnetometer at different temperatures. The out-of-plane magnetic coercivity of the films was found to increase with film thickness and then decreases with further increase in thickness. Maximum coercivity of ~ 20 kOe has been obtained for the 400 nm thick film. Magnetic domain patterns were studied using magnetic force microscopy (MFM) technique and the observed magnetic properties are correlated with domain pattern and microstructures.

Although there are several reports on device applications of Sm-Fe thin films which exhibit negative magnetostriction, a comprehensive study on the effect of different process parameters on the magnetic properties and its correlation with structure and microstructure is still elusive. Hence, Sm-Fe films were deposited on Si (100) substrate using dc magnetron sputtering technique under varying Ar pressure and sputtering power. Effect of these parameters on the microstructural and magnetic properties of the films was studied in detail and is presented in chapter 5. The curie temperature of the films was found to increase with increase in sputtering power and Ar pressure. This was attributed to increase in film thickness and size of islands (atomic clusters). Coercivity as low as 30 Oe has been achieved in the film deposited at 15 mTorr Ar pressure. The Curie temperature for the films deposited at higher Ar pressure (10 and 15 mTorr) are found to be above RT. Maximum saturation magnetostriction of ~ - 390 μ-strains has been achieved in the film deposited at 15 mTorr Ar pressure. Rapid thermal processing (RTP) experiments were also carried out to increase the magnetic ordering in the films deposited at low Ar pressure (5 mTorr) by imparting structural ordering. Large improvement in magnetization and Curie temperature of the film was observed after RTA. However, this could be attributed to the formation of nano-crystalline Fe phase as evidenced from the TEM studies and thermomagnetic measurements.

An overall summary of the experimental results has been presented in chapter 6. The scope of work for further study in future has also been highlighted in chapter 7.